Assessment of fissionable material behaviour in fission chambers

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OUTLINE

PART I: Introduction

PART II: Computation of the reaction rates in a fission chamber and its sensitivity to neutron spectrum

- Reaction rates and saturation current delivered by a fission chamber
- Sensitivity of the saturation current with respect to the neutron spectrum

PART III: Other parameters having influence on the fission chamber behaviour

- Total radioactivity
- Xenon inventory
- Temperature effect
- Deposit thickness

PART IV: Impact of activation cross section uncertainties

CONCLUSIONS

INTRODUCTION

- **A comprehensive study is performed in order to assess the pertinence of fission chambers coated with different fissile materials for high neutron flux detection**

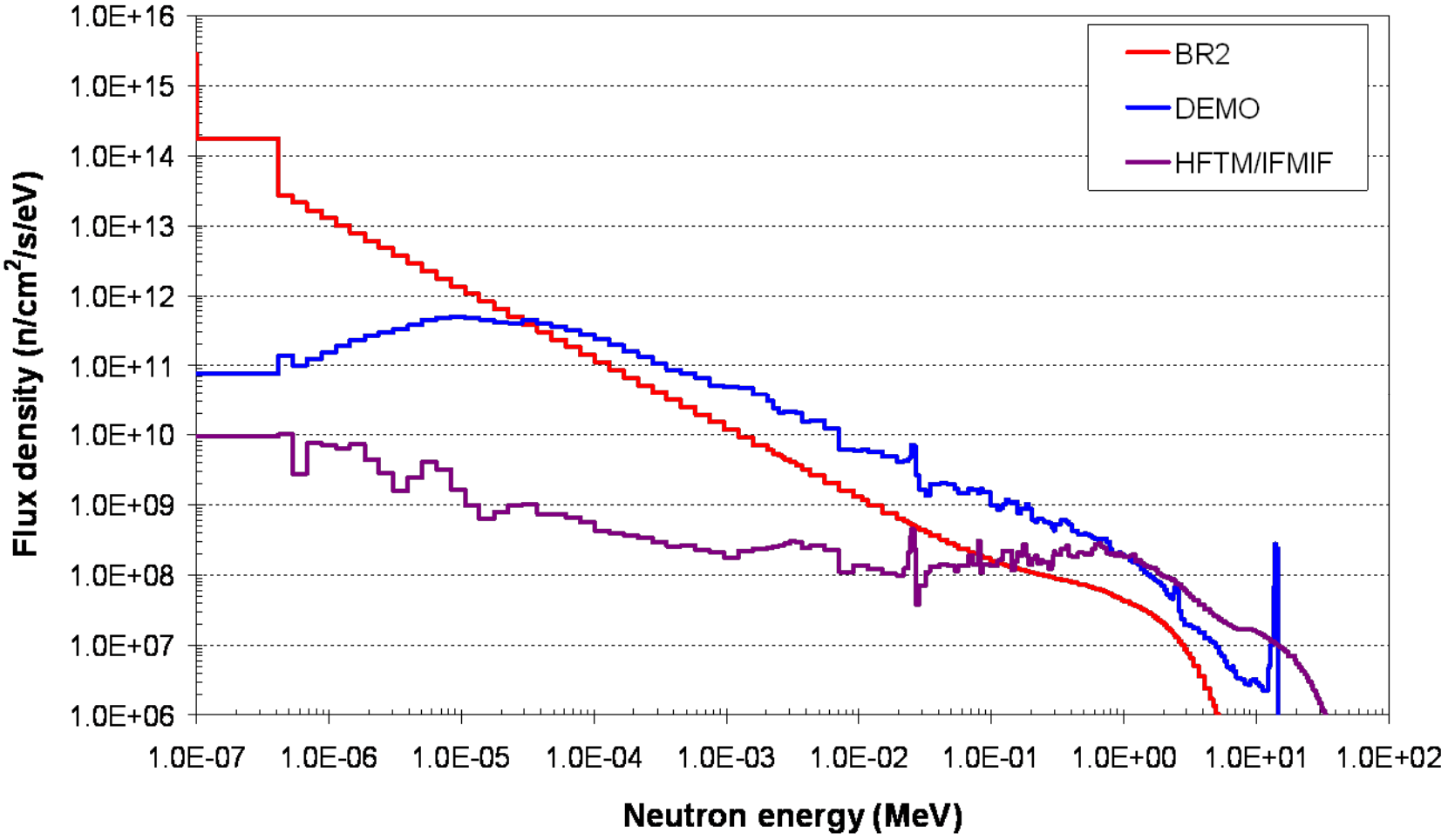
- **Three neutron scenarios are proposed to study the fast component of a high neutron flux:**
  - **High neutron flux with a significant thermal contribution such as BR2**
  - **DEMO magnetic fusion reactor**
  - **IFMIF high flux test module**

- **In this study, the inventory code ACAB (**NEA-1839**) is used to analyze the following questions:**
  - **Impact of different deposits in fission chambers**
  - **Effect of the irradiation time/burn-up on the concentration**
  - **Impact of activation cross section uncertainties on the composition of the deposit for all the range of burn-up/irradiation neutron fluences of interest**

- **The complete set of nuclear data (**decay, fission yield, activation cross sections and uncertainties**) provided by EAF2007 data library are used for this evaluation.**

PART I: Introduction: Neutron scenarios

Figure 1. Neutron spectra of different neutron environments in a VITAMIN-J+ (211) multi-group structure.



PART II: Reaction rates and sensitivity to neutron spectrum

Table I. Total neutron flux and fraction of neutrons in four energy ranges for the three neutron scenarios.

	% neutrons having energies				Total neutron flux (x 10^{14}) ($n/cm^2/s$)
	E <0.625eV= E_0 *thermal*	E_0< E <1 MeV *epithermal*	1 MeV < E <20 MeV *fast*	E >20 MeV *extended*	
BR2	55.53	35.99	8.48	-	6.16
DEMO	0.00	66.72	33.28	-	13.0
IFMIF/HFTM	0.00	27.28	66.53	6.19	7.30

The total neutron flux can be decomposed into four components as follows:

$$\phi_{total} = \sum_g \phi_g = \sum_{I=ther,epi,...} \left(\sum_{g \in G_I} \phi_g \right) = \sum_I \phi_I = \phi_{thermal} + \phi_{epi} + \phi_{fast} + \phi_{extended}$$

The collapsed one group cross section with a multigroup neutron spectrum is written as:

$$\frac{\sum_{g \in G_I} \sigma_g \phi_g}{\sum_g \phi_g} = \frac{\sum_{g \in G_I} \sigma_g \phi_g}{\phi^{total}} = \overline{\sigma}_I$$

The total fission reaction rate for a given neutron spectrum and for *a given fissile deposit target* can be computed as:

$$R_{tot}(t) = \sum_i N^i(t) \overline{\sigma}^i_{fiss} \phi_{total}$$

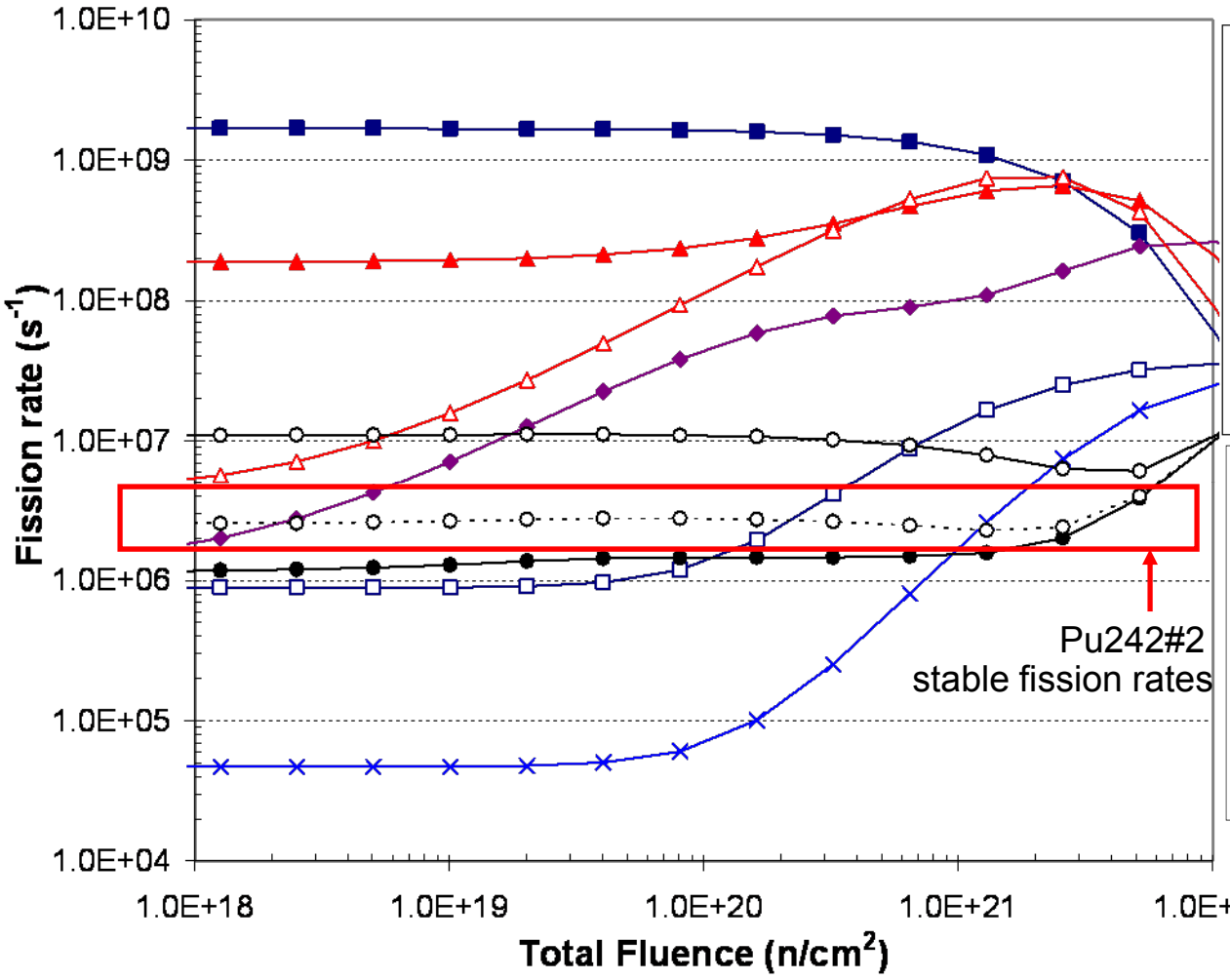
PART I: Introduction: Other realistic fissile deposits?

Table II. Isotopic composition of the potential fissile deposit or solutions.

Deposit	Mass (μg)	Atomic percentage in %									
		^{232}Th	^{235}U	^{238}U	^{237}Np	^{238}Pu	^{239}Pu	^{240}Pu	^{241}Pu	^{242}Pu	^{244}Pu
Th232	3.85	100									
U235	3.90		97	3							
U238	3.95		0.04	99.96							
Np237	3.93				100						
Pu238	3.95					94.95	4.58	0.40	0.02	0.05	
Pu240	3.98					0.02	0.08	99.88	-	0.02	
Pu242	4.02									100	
Pu242 #1	4.02					0.214	0.116	0.172	0.180	99.274	0.044
Pu242 #2	4.02					0.004	0.005	0.022	0.035	99.932	0.002

PART II: Reaction rates

Figure 2. Fission rates (fission/s) for the different pure and solution deposits in a typical high flux thermal neutron environment (BR2).



The total fission reaction rates are directly related with the current delivered by the FC:

$$I_{sat} = f(L, p, r_c, r_a, ...) \cdot \sum_{i=1}^{ITOT} N^i(t)\, \bar{\sigma}^i_{fission}\ \phi_{total}$$

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PART II: Sensitivity to neutron spectrum

We define the relative change in the saturation current (under flux variations) as:

$$\frac{\Delta I_{sat}}{I_{sat}} = \sum_{I=ther,epi,...} \left(\frac{\partial I_{sat}}{\partial \phi_I}\bigg|_{\phi \neq \phi_I, cte} \frac{\phi_I}{I_{sat}} \right) \frac{\Delta \phi_I}{\phi_I} = \sum_{I=ther,epi,...} S_I \frac{\Delta \phi_I}{\phi_I}$$

The sensitivity coefficient of the current with respect to a flux variation is

$$S_I = \frac{\partial I_{sat}}{\partial \phi_I}\bigg|_{\phi \neq \phi_I, cte} \frac{\phi_I}{I_{sat}} = \left(\sum_i N^i \hat{\sigma}_I^i \right) \frac{\phi_I}{R_{tot}} = \frac{R_I}{R_{tot}} \qquad \text{with:} \quad \sum_I S_I = 1$$

The fission reaction rates in each energy-region can be defined by

$$R_I = \sum_i N^i \left(\sum_{g \in G_I} \sigma_g^i \phi_g \right) = \sum_i N^i \hat{\sigma}_I^i \phi_I = \sum_i N^i \overline{\sigma}_I^i \phi_{total}$$

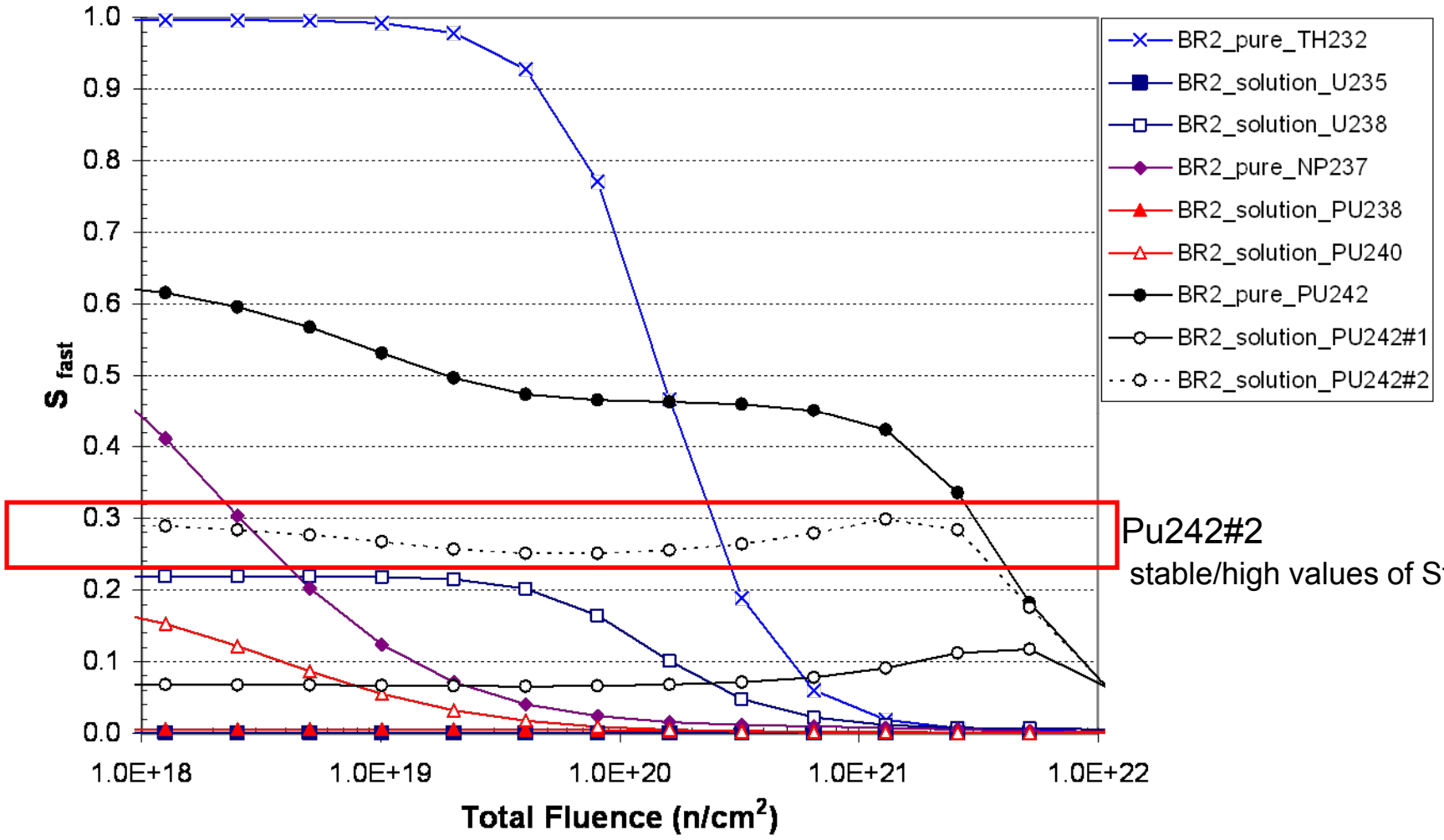
In the case of an impure deposit, we define S_I as: $S_I = \sum_k \alpha_k S_I^k \beta_k$

where:

- α_k is the atomic percentage of the pure deposit *k*
- S_I^K is the sensitivity of the pure deposit k
- β_k is R^k/R_{tot}, the relative fraction of fission rate of the pure deposit k

PART II: Sensitivity to neutron spectrum

Figure 4. Sensitivity to fast neutrons for initially different pure and solution deposits in a typical high flux thermal neutron environment (BR2).

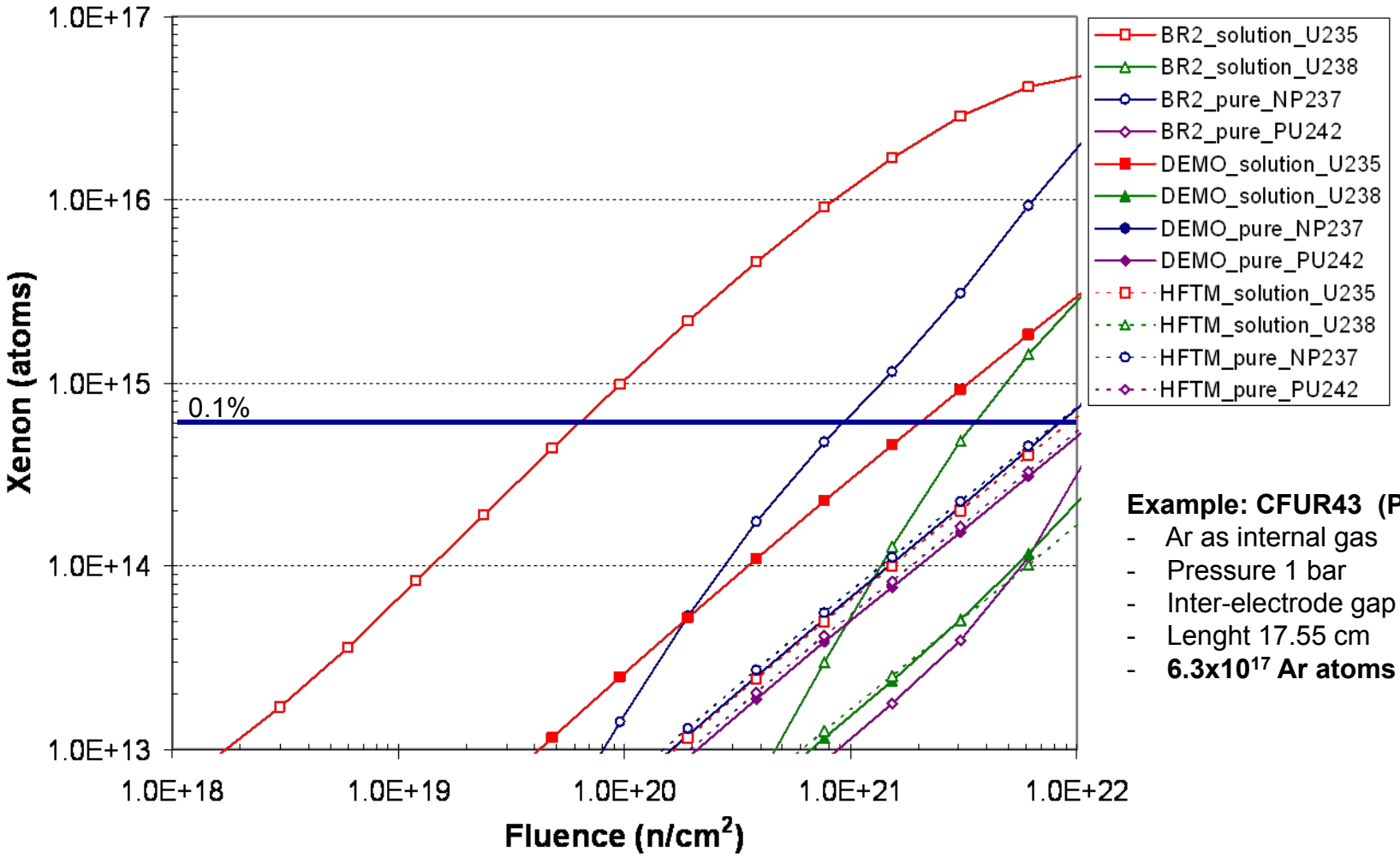


PART III: Other parameters having influence in FC behaviour

- **Total radioactivity**
  - **Evolution of total activity induced in the FC (waste management)**
- **Xenon inventory**
  - **Addition of contaminants with low ionization potential (e.g. Xenon ~ 0.1%)**
- **Deposit thickness**
  - **To analyze the self-shielding phenomenon:** $\delta^{\max} < \varepsilon / \Sigma_{total}$
  - **To assess the fission product trapping within the deposit (auto-absorption)**
- **Temperature effect**
  - **We assess the temperature effect for different deposits irradiated in BR2**
  - **Two sets of calculations have been performed:**
    - **"BRANCHING" cases**, the reference temperature of 325K used is instantaneously changed to 350K at each irradiation time. Assuming a constant total neutron flux, the differences in the total fission rates are due to the different one-group fission cross sections (each one collapsed with a different spectrum).
    - **"SPECTRAL HISTORY" cases**, "SH", we keep the temperature at 350K during the whole irradiation time, so not only the one-group fission cross sections change, but also the atomic concentration.

PART III: Xenon

Figure 5. Contamination of the filling gas by Xenon (atoms) induced in the fission chamber for initially different pure (Np237 and Pu242) and solution (U235 and U238) deposits in typical high neutron flux environments

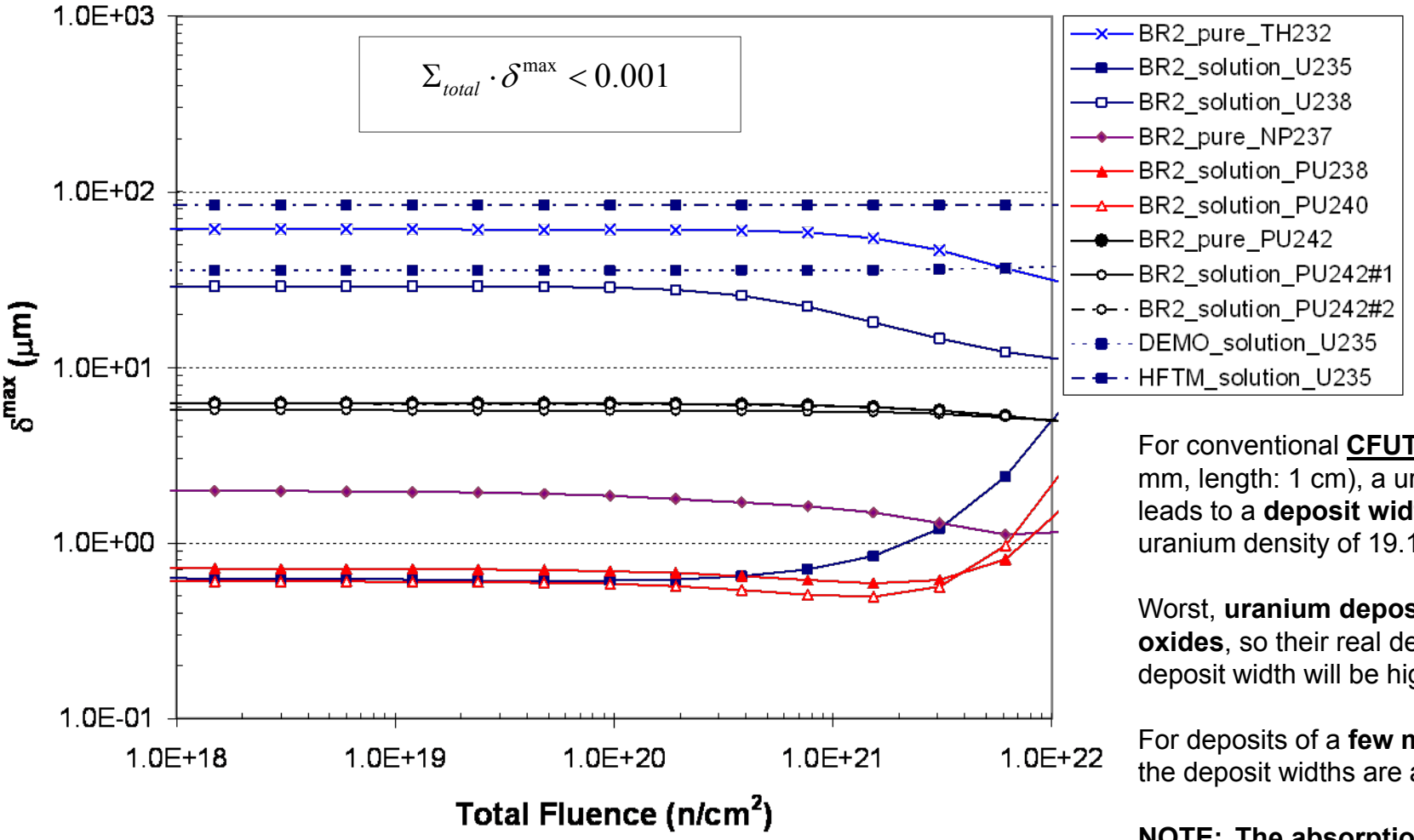


Example: CFUR43 (PHOTONICS)

- Ar as internal gas
- Pressure 1 bar
- Inter-electrode gap 0.25 mm
- Lenght 17.55 cm
- **6.3×10^{17} Ar atoms**

PART III: Self-shielding phenomenon

Figure 6. Predicted maximum thickness assuming an interaction probability (ε) of 0.001



For conventional **CFUT-C3** anodes (radius: 1.25 mm, length: 1 cm), a uranium deposit of **3 g** leads to a **deposit width of 1.3 mm**, with a uranium density of 19.1 g/cm3 .

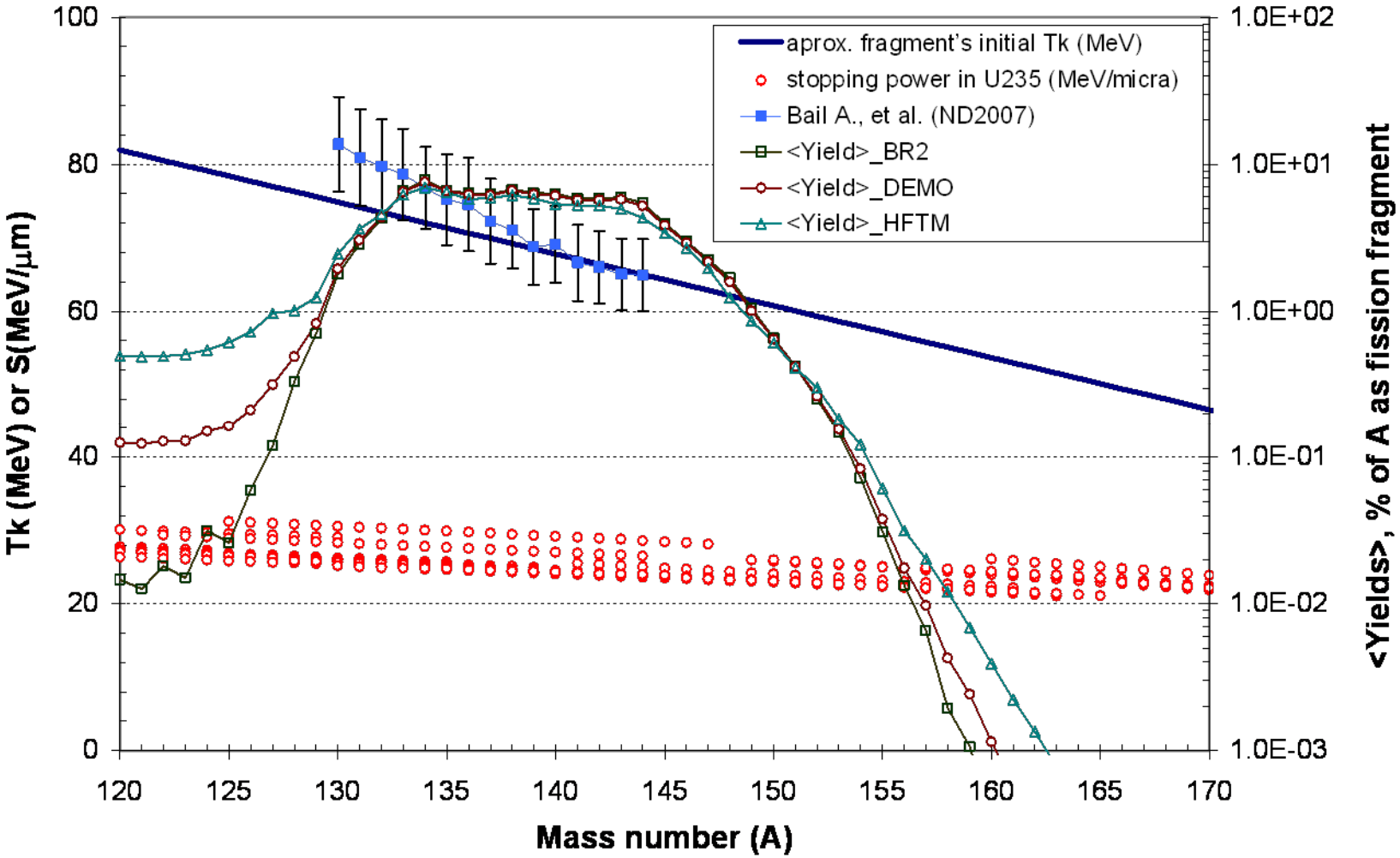
Worst, **uranium deposits are essentially oxides**, so their real density is lower and the deposit width will be higher.

For deposits of a **few micrograms**, however, the deposit widths are around **1 nm**.

NOTE: The absorption of FPs within the deposit could become non negligible for large deposits of a few grams.

PART III: Fission product trapping within the deposit

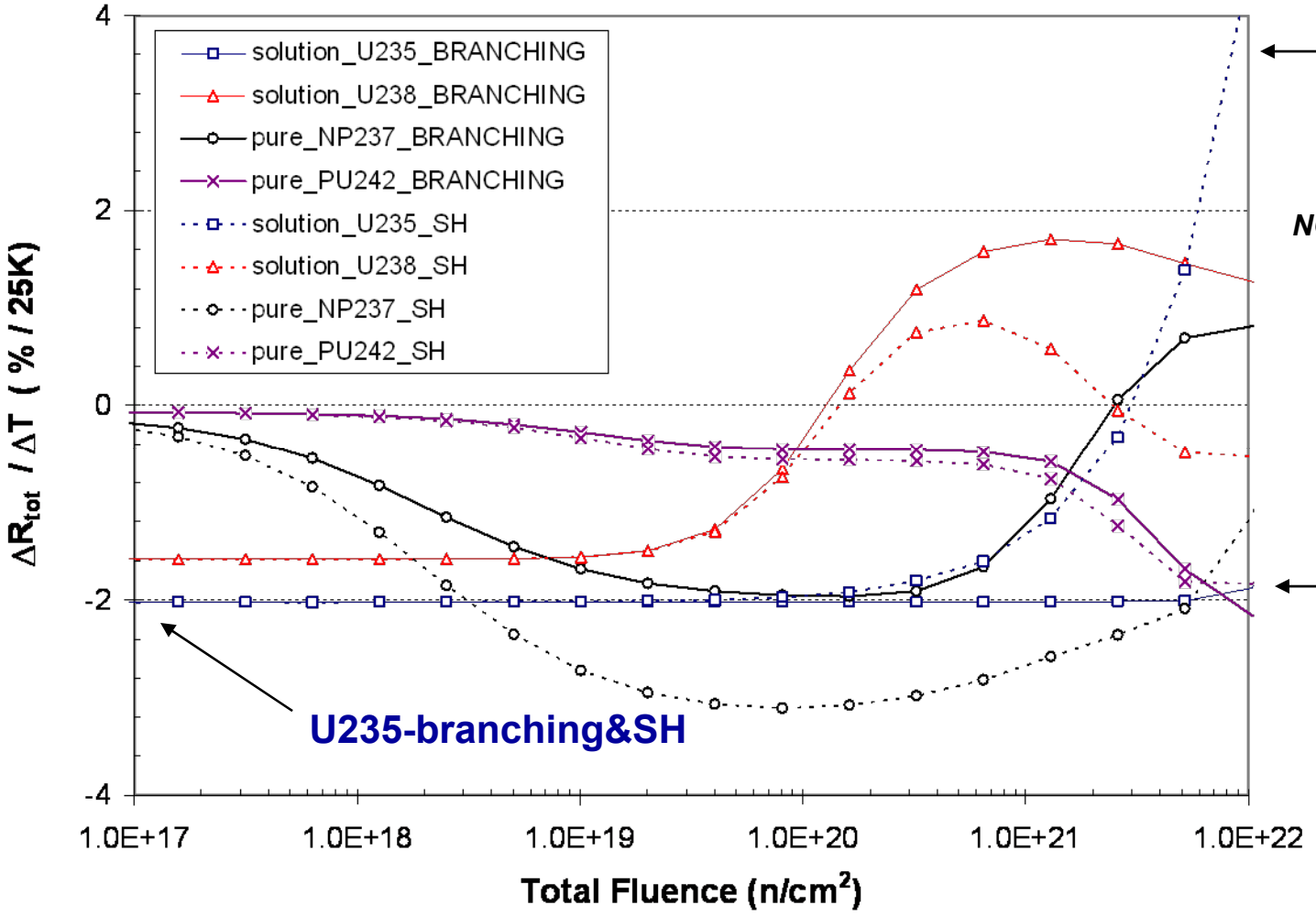
Figure 7. On the left axis, the roughly total kinetic energy of any fragment with mass number A, mean and relative error kinetic energy (one standard deviation) from fission of $^{235}U(nth,f)$ taken from [14] and the stopping power (MeV/μm) at this energy calculated with SRIM in Uranium for different elements (charge Z) with the same mass number.



On the right axis, the effective fission product yield for ^{235}U collapsed by ACAB code for different neutron spectra.

PART III: Temperature effect: Branching/Spectral History cases

Figure 8. Temperature dependence of the fission rates for different deposits.



NOTE: **temperature effects** should be studied more carefully by experimentalists, especially **in thermal reactors** having significant temperature gradients.

- **Branching cases** show the difference in % between a reference irradiation case at 325K and an instantaneous change in the temperature of +25K
- **Spectral history cases**: we compare the nominal case at 325K with a modified irradiation case at 350K

PART IV: Impact of activation cross section uncertainties

Table III. Total relative experimental error (in %) for capture and fission cross sections from EAF2007/UN.

	Fission			Capture		
	BR2	**DEMO**	**HFTM**	**BR2**	**DEMO**	**HFTM**
Th232	16.6	16.7	13.7	2.5	3.8	15.4
U235	3.1	7.0	15.1	3.0	4.8	15.5
U238	16.6	16.7	13.9	2.8	3.3	13.5
Np237	15.8	16.6	15.4	3.2	7.6	15.0
Np238	46.7	24.7	30.7	33.0	24.5	32.3
Pu238	5.4	12.5	15.6	3.9	10.0	15.9
Pu239	3.3	6.0	15.3	4.5	8.6	14.3
Pu240	14.0	16.2	14.9	2.5	3.5	7.8
Pu241	3.3	8.8	15.1	3.0	8.0	16.3
Pu242	14.8	16.5	15.2	8.0	8.0	14.0
Pu243	118.4	47.67	59.5	274.6	23.6	32.4
Pu244	16.5	16.6	15.2	15.1	23.9	32.6

Given ***V*** the G-by-G variance matrix of the relative cross sections vector, the variance Δ^2 of the relative spectrum-averaged cross section is:

$$\Delta^2 = \omega^T V \omega$$

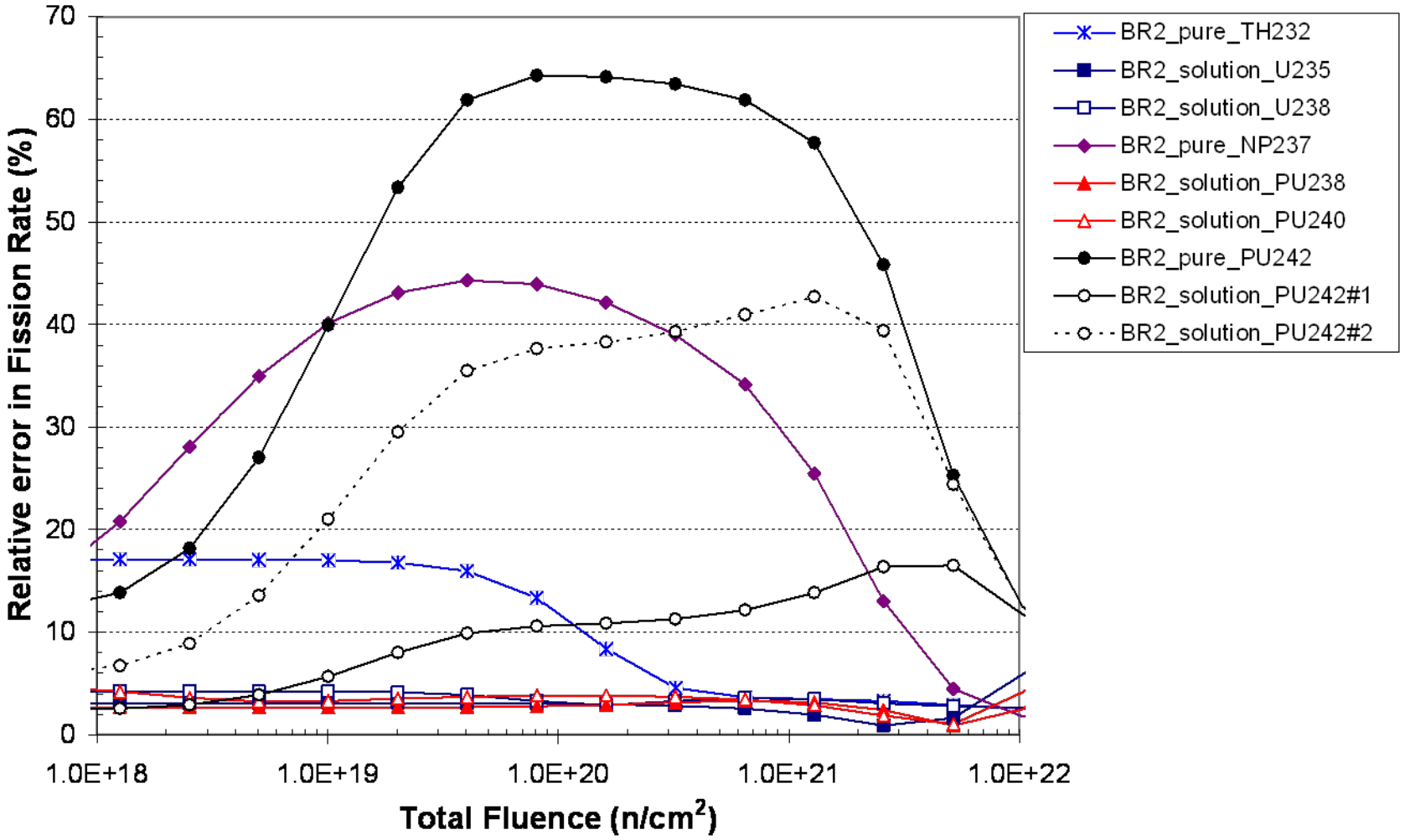
with: $\omega = [\frac{\phi_1}{\bar{\phi}}\frac{\sigma_1}{\sigma^{eff}}, \cdots, \frac{\phi_G}{\bar{\phi}}\frac{\sigma_G}{\sigma^{eff}}]^T$

the total flux as: $\bar{\phi} = \phi_1 + \phi_2 + \cdots + \phi_G$

and: $\sigma^{eff} = \frac{\phi_1\sigma_1 + \phi_2\sigma_2 + \cdots + \phi_G\sigma_G}{\phi_1 + \phi_2 + \cdots + \phi_G}$

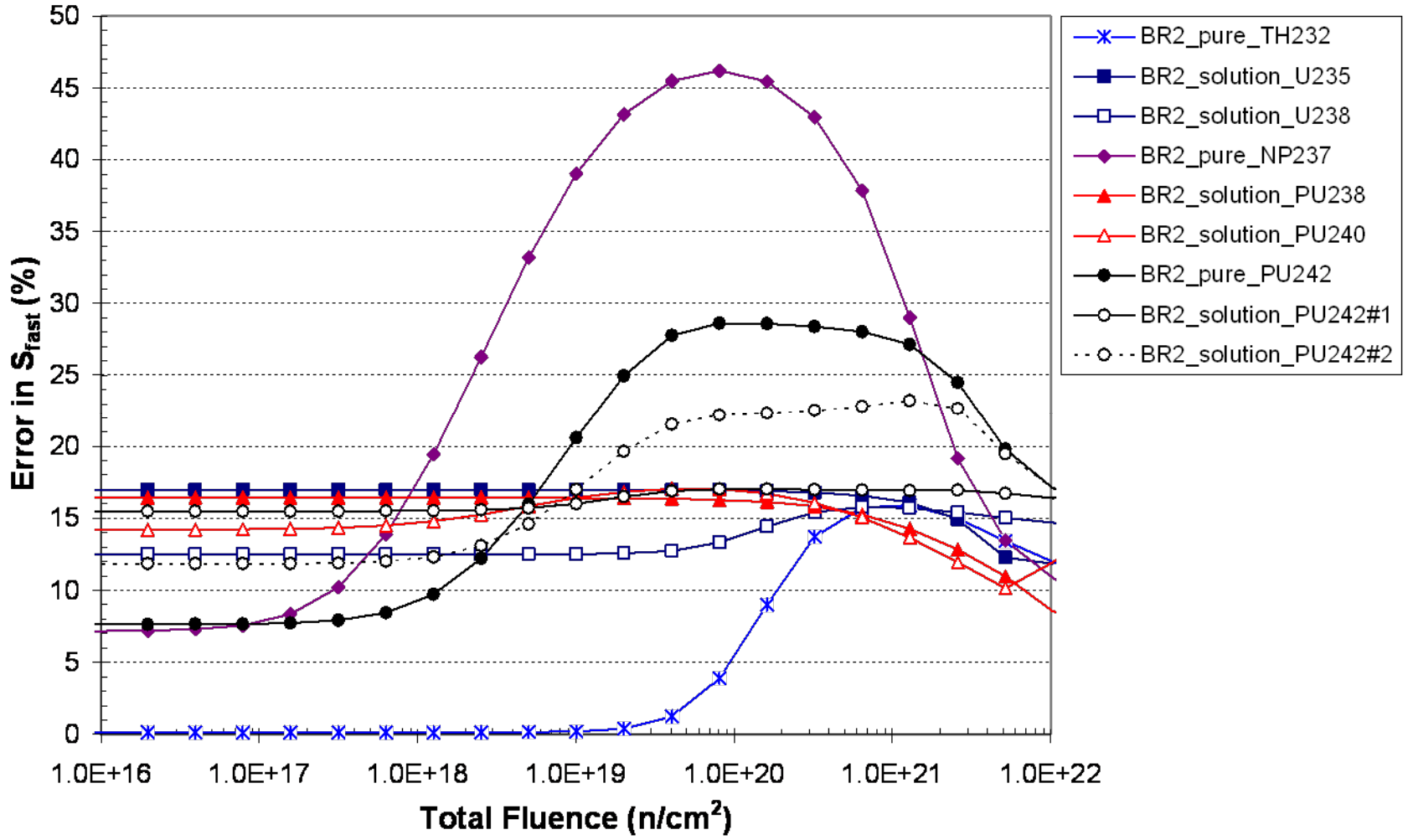
PART IV: Relative error in fission rates in BR2

Figure 9. Relative error in fission rates (in %) for different pure and solution deposits in a typical high flux thermal neutron environment (BR2).



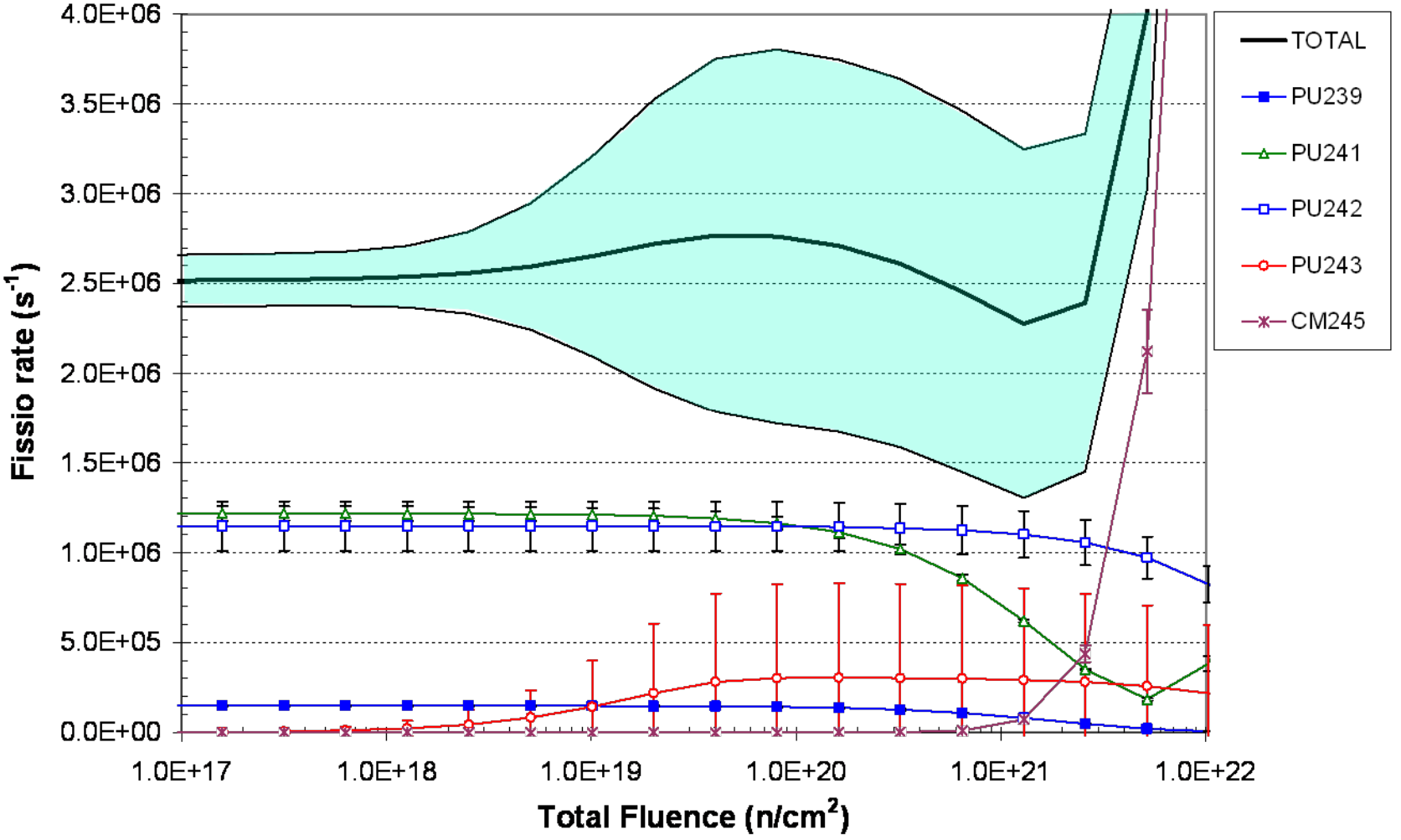
PART IV: Relative error in Sfast in BR2

Figure 10. Relative error in Sensitivity to fast neutrons (in %) for different pure and solution deposits in a typical high flux thermal neutron environment (BR2).



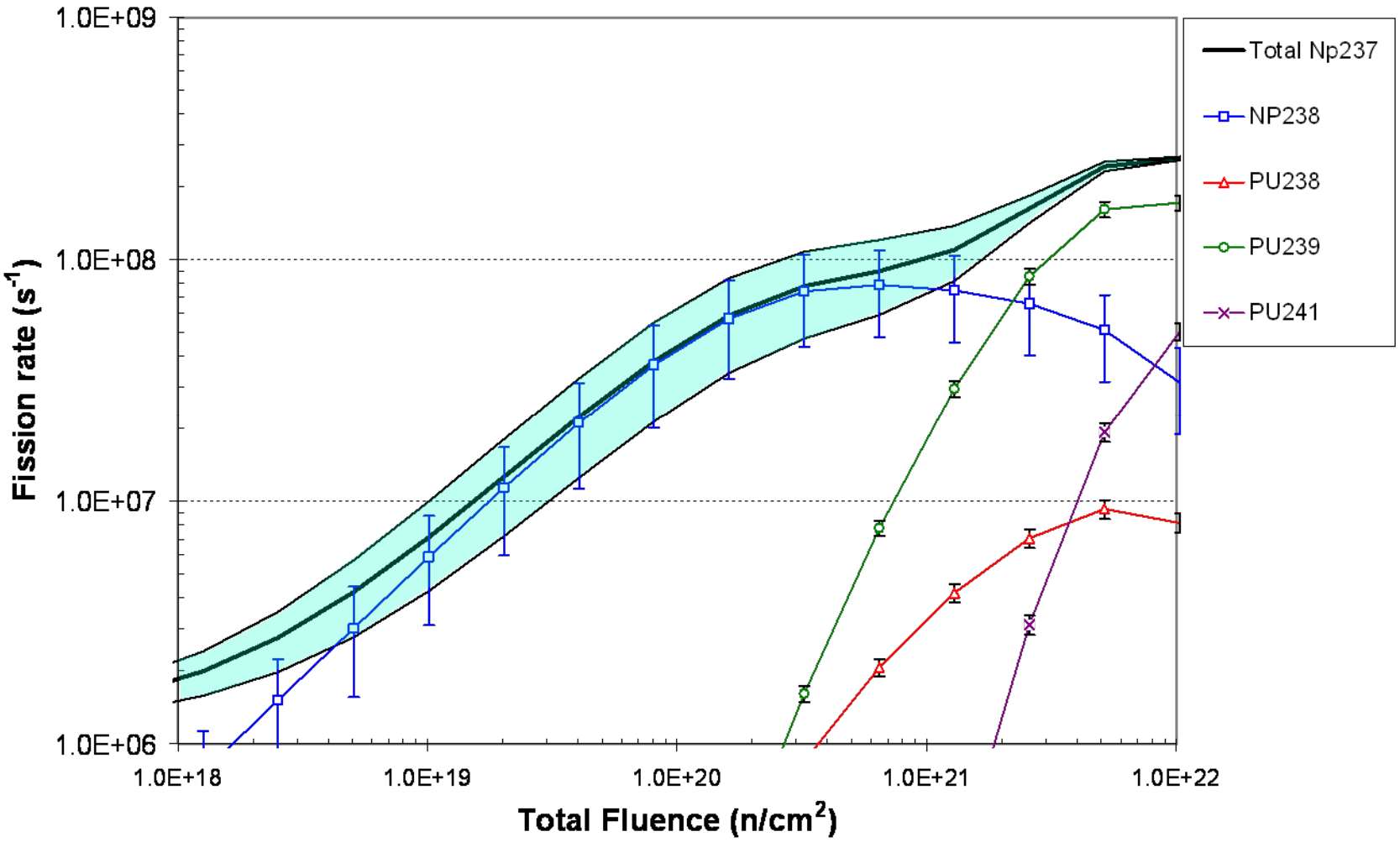
PART IV: Relative error in fission rates Pu242#1 in BR2

Figure 11. Contribution and error bars (**one standard deviation**) of each isotope in the total fission rate for a deposit of Pu242#2 (see Table I for initial composition) in a typical high flux thermal neutron environment (BR2).



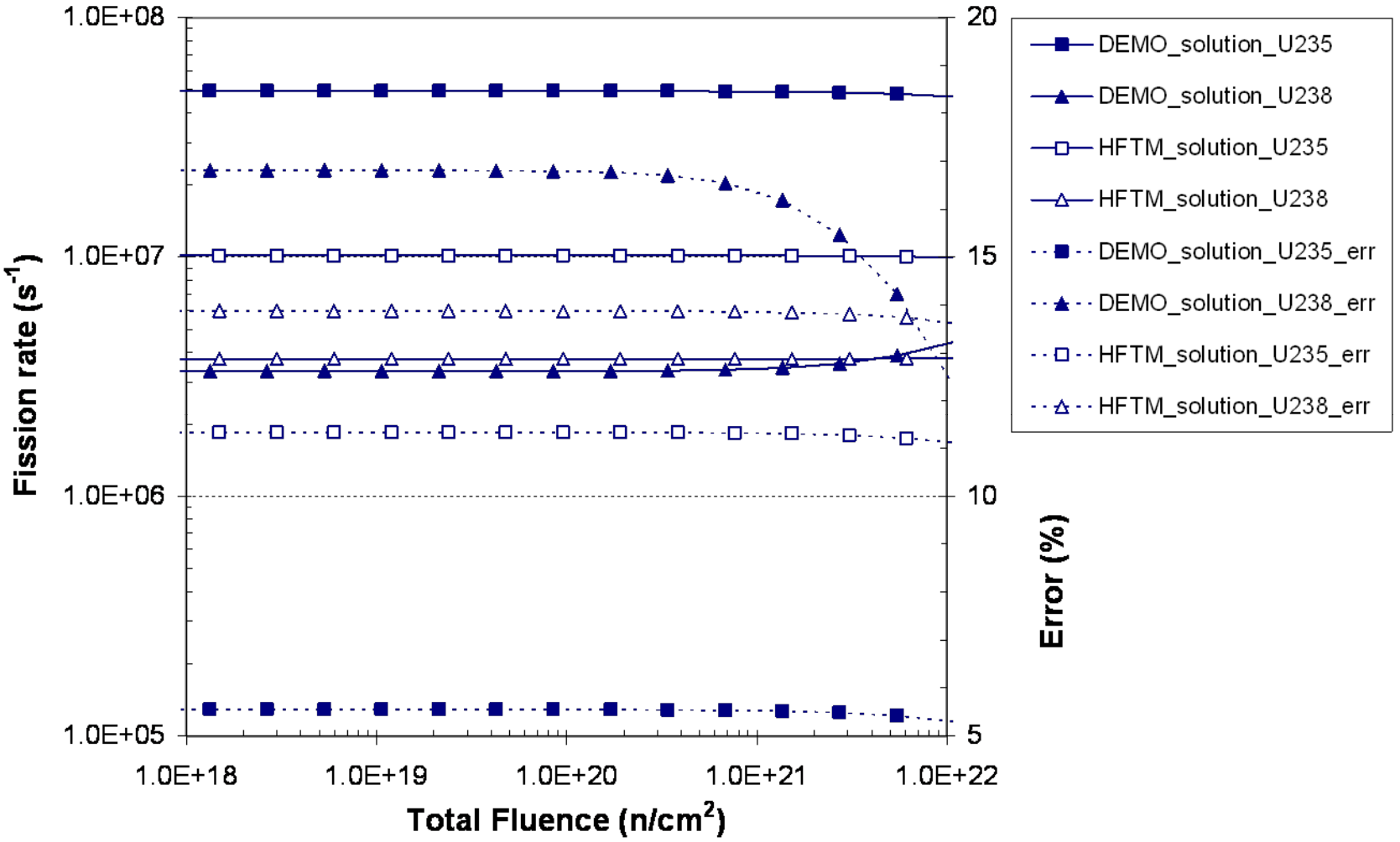
PART IV: Relative error in fission rates Np237 in BR2

Figure 12. Total fission rate and error bars (**one standard deviation**) for initially pure Np237 deposit irradiated in a high flux thermal environment (e.g. BR2). Contributions of each isotope to the total fission rate and errors are shown.



PART IV: Relative error in fission rates DEMO/IFMIF

Figure 13. Fission rates and relative error (in %) for different U235 and U238 deposits in DEMO and HFTM/IFMIF neutron environments.



CONCLUSIONS

- **We present an assessment of fissionable material behaviour in three neutron scenarios with different degrees of hardness (BR2, DEMO and IFMIF)**
  - **The evolution of fission rates as a function of the fluence for some potential/realistic deposits or solutions are predicted as well as other parameters having influence on the FC behaviour for a long-term performance (sensitivities to fast neutrons, xenon prediction and spectral history effect in BR2 due to changes in temperature)**
    - In BR2, the fission rates are stable with deposits of Pu^{242}#1,2 up to fluences as high as 10^{22} n/cm^2 as well as satisfying high values of S_{fast}
    - For DEMO and IFMIF, fission rates remain stable for the complete set of deposits

- **Uncertainty calculation due to uncertainties in activation cross sections (a Monte Carlo technique implemented in ACAB code has been used to propagate ND uncertainties)**
  - **In BR2, large uncertainties were found in deposits of Np237 and Pu242 due to uncertainties in fission cross sections of ^{238}Np and ^{243}Pu, respectively**
  - **In addition, we have found that the uncertainty in the contribution of ^{245}Cm at high fluences in the deposits of Pu242 is mainly due to the uncertainty in its inventory**
  - **For other deposits, uncertainties remain below 5% for fission rates up to fluences of 10^{22} n/cm^2.**
  - **In the HFTM/IFMIF and DEMO, we found relative errors in fission rates between 5% and 17%**

  - **In conclusion, the knowledge of the evolution of these uncertainties can help to better understand the expected responses of fission chambers**